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Municipal wastewater treatment by modified tannin flocculant agent

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ABSTRACT

A new tannin-based coagulant and flocculant agent (TANFLOC) has been tested in order to treat urban wastewater. TANFLOC has showed a high effectiveness in turbidity removal (almost 100%, depending on the dosage) and around 50% of BOD_5 and COD removal, which makes TANFLOC an appropriate coagulant agent with an efficiency that is comparable to alum's. Coagulant and flocculant process does not depend on temperature, and optimum agitation speed and time have been found to be 40 rpm for 30 min. Polyphenol content does not increase drastically, and 30% of anionic surfactants are removed. Sedimentation process seems to be a flocculent separation so Sludge Volumetric Index and its evolution with flocculant dosage have been determined. TANFLOC has been revealed as a quite effective coagulant and flocculant agent in wastewater treatment.

1. Introduction

Human activity is a source of wastes. Particularly in urban settlements, wastewater that came from domestic and industrial effluents may be a hazardous, noxious product [1] which should be adequately treated in order to avoid environmental [2] and health implications [3,4]. 2008 has been actually declared the International Year of Sanitation by the General Assembly of the United Nations through its Resolution A/C.2/61/L.16/Rev.1 dated on December, 4th, 2006. Ineffective sanitation infrastructures facilitate every year 2.2 millions of deaths by diarrhoea, mainly among child under 3 years old, 6 million people blind from *trachoma* and 200 million people infected with *schistosomiases*, just for giving some data [5]. Obviously, most of them in developing countries, so appropriate technologies referring urban wastewater may be investigated in order to broaden the variety of technical possibilities of treatment.

In this sense, many types of water treatment are being used. Their differences lay on economical and technical features. Some interesting papers have been published about several natural and alternative ways of municipal wastewater treatment involving green filters [6] chemical primary separation and UV disinfection [7] or multi-stage procedures [8] in order to get rid of dangerous pollution.

Several previous papers have pointed out the importance of urban wastewater management [9,10]. This type of waste has been a target for social studies, as it involves several aspects that have to do with social structure and community organization [11,12]. According to this dimension, it is very important to consider wastewater management as a social change factor in developing countries, as the relationship between wastewater treatment and production, on one hand, and human developing process, on the other, is rather known [13].

Researching on other procedures of water treatment has been the scope of this and other papers. For several years, investigators are concerned towards cooperation among developing countries and they are working on an alternative process for water treatment, mostly bearing in mind concepts such as sustainability, affordability and social feasibility. In this sense, natural coagulants/flocculants are wide-spread, easy-handling resources that are not difficult to work with by non-qualified personnel. There are some examples of this agent, such as *Moringa oleifera* [14] or *Opuntia ficus* [15]. Tannins may be a new source for coagulant and flocculant agents.

Few authors have investigated about tannin water treatment capacity.

Özacar and Sengil [16] characterized tannins obtained from *valonia*, an autoctonous tree from Turkey, and used them for coagulation–flocculation process of wastewater. The authors demonstrated that tannin has a very good effect, combined with $Al_2(SO_4)_3$ in order to enhance further stages of sludge removal.

Zhan and Zhao [17] tried to remove lead from water by using an adsorbent, tannin-based gel. Process of metal removal is improved by tannin gelification. In the same sense, there are other references such as Nakano et al. [18] and Kim and Nakano [19].

Özacar and Sengil [20] enhanced the previous article and gave special data about trihalomethane formation and other undesirable compounds, as well as treated water safety. They worked always with tannin–$Al_2(SO_4)_3$ combination.

Palma et al. [21] used tannins extracted *in situ* from *Pinus radiata* bark in order to polymerize a solid which is used in heavy metal removal. Bark itself was combined with a tannin solid into adsorption columns.

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TANFLOC flocculant product is a trademark that belongs to TANAC (Brazil). It is a tannin-based product, which is modified by a physico-chemical process, and has a high flocculant power. It is obtained from an *Acacia mearnsii* bark. This tree is very common in Brazil and it has a high concentration of tannins.

According to TANAC specification, TANFLOC is a vegetal water-extract tannin, mainly constituted of flavonoid structures with an average molecular weight of 1.7 kDa. More groups such as hydrocolloid gums and other soluble salts are included in the TANFLOC structure. Chemical modification includes a quaternary nitrogen that gives TANFLOC cationic character.

Several references have been found regarding this kind of chemical processes [22–24]. Most of them are patents, including the specific process for TANFLOC, which is reported [25]. The scientific literature refers a reaction mechanism that involves three reagents: a tannin mixture, mainly polyphenol tannins whose structure may be similar to flavonoid structures such as resorcinol A and pyrogallol B rings; an aldehyde such as formaldehyde and an amino compound, such as ammonia or a primary or secondary amine or amide compound [26]. The three reagents, under certain conditions of pH (under 7) and temperature (80 °C), may produce the mentioned flocculant agents.

Under *tannin* denomination there are lots of chemical families. Tannins have been used traditionally for tanning animal skins, but it is possible to find several products that are distributed as flocculants. Tannins come from vegetal secondary metabolites [23]: bark, fruits, leaves… *Acacia* and *Schinopsis* are well-known tannin feedstock. However, it is not needed to search for tropical species: *Castanea*, *Quercus ilex*, *suber* or *robur* have also tannin-rich bark.

The main aim of the present investigation is to characterize the coagulant and flocculant activity of this new tannin-based product as a municipal wastewater treatment. The chemical modification made on *A. mearnsii* tannin is not quite difficult and it is widely reported as Mannich base reaction [22], although specific industrial process for TANFLOC is under intellectual patent law. Therefore, this investigation should be considered as an initial approach to these kinds of coagulant and flocculant agents.

2. Materials and methods

2.1. Reagents

TANFLOC has been kindly supplied by TANAC (Brazil). $Al_2(SO_4)_3.18H_2O$ has been supplied by SIGMA. All reagents involved in analytical procedures have analytical purity.

2.2. Raw water

Raw water was obtained from the Wastewater Treatment Plant of La Albuera, a little town near Badajoz (South West of Spain). This treatment plant was designed some years ago. It receives municipal wastewater from 4000 people. There are no significant influents of industrial wastewater, but some agricultural and livestock farms are present, so such diffuse pollution may occur. The effluent has a moderately low COD charge. Average incoming flow rate is 41.63 m^3/h. Water involved in this study is collected after previous big solids separation and before oil and sand separation. The main physico-chemical characteristics of this water are referred in Table 1. If compared with other wastewater data found in literature [8,27] our working water has less pollutant charge, due surely to the nature of dumpings and above all the domestic origin of wastes.

2.3. Jar-test procedure

Jar-test was selected as the standard treatment in order to study flocculant process [28]. The procedure was: 0.5 L of turbidity-known wastewater was put into a beaker. Certain dose of flocculant was added, and beaker was put into a Jar-test apparatus (VELP-Scientifica JLT4). Two stirring periods were applied: one at 100 rpm for 2 min and another one at a lower speed for a longer period. In order to study the influence of this last period, its duration and agitation intensity were varied. Turbidity was measured by a HI93703 turbidimeter (Hanna Instruments) 1 h after Jart-test was finished. Turbidity sample was obtained from the center of the beaker, 3 cm from the surface.

Table 1
Raw water characterization data.

Parameter	Value	Units
Turbidity	82.5	NTU
Suspended solids	100	ppm
Total solids	650	ppm
Anionic surfactants	3.9	ppm
Polyphenols	6.4	Tannic acid equivalent ppm
$KMnO_4$ oxidability	65.6	O_2 ppm
Biological oxygen demand	130	O_2 ppm
Chemical oxygen demand	210	O_2 ppm
Chloride	21.3	Cl^- ppm
Calcium	94.6	Ca^{2+} ppm
Hardness	444	$CaCO_3$ ppm
Conductivity	1006	µS cm^{-1}
Nitrate	22.5	NO_3^- ppm
Nitrite	0.04	N ppm
Ammonium	2.1	N ppm
Phosphate	7.3	P ppm
Total phosphorus	11.9	P ppm
pH	8.2	

2.4. Analytical methods

All analytical measures were made according to the American Public Health Association standard methods [29]. Measures referring sludge production and Sludge Volumetric Index (SVI) were done with a 25-mL calibrated test tube and 1-L Imhoff cone. In the first case, a 25-mL sample was collected just after coagulation and flocculation process (without sedimentation) and suspended solids were determined by *millipore* fine filtration (45 µm glass fiber filter). In the second case, Imhoff cone received a 0.5-L sample of treated water and it was allowed to settle for 1 h. Then, sludge volume was measured as Imhoff cone was calibrated.

Anionic surfactants were determined by a method based on methylene blue-anionic surfactant association [30]. 10 mL of clarified sample was put into a separation funnel. 25 mL of trichloromethane (PANREAC) and 25 mL of methylene blue solution (PANREAC) were added and the funnel was shaken vigorously. Organic fraction was taken out and put into another separation funnel, in which 50 mL of cleaning solution was added. Funnel was shaken again, and the resultant organic fraction was put into a 25-mL flask. It was filled up to the mark with trichloromethane and surfactant concentration was determined by visible spectrophotometry at 625 nm, with zero made with pure trichloromethane by using a HEλIOS spectrophotometer.

Reagents were prepared in the following way:

- *Cleaning solution*: 43.5 g of NaH_2PO_4 (ALDRICH) was taken and it was diluted into 500 mL of distilled water. 6.6 mL of H_2SO_4 (PANREAC) 98% w/v was added and dilution was raised up to 1 L.
- *Methylene blue solution*: 30 mg of methylene blue (ALDRICH) was added to 1 L of cleaning solution.

Calibration equation was done with sodium lauryl sulphate (PANREAC).

Polyphenol concentration was determined by *Folin-Ciocalteau* test [31]. 20 mL of sample was put into a 50-mL flask. 2.5 mL of Folin-Ciocalteau's phenol reagent (FLUKA) was added. Then, 5 mL of 20% sodium carbonate solution (SIGMA) was added, and the flask was filled up to the mark with distilled water. Absorbance was measured at 725 nm after 1 h. Zero was made with reagents without sample in a

50-mL flask, filled up with distilled water. Results are expressed in tannic acid equivalent ppm.

For the COD determination, a Selecta Tembloc oven mod was used. A PF-10 Macherey-Nagel photometer, and test cuvettes were pre-prepared for the desired measurement range (the range of concentrations selected was 50–1000 mg O_2/L). In addition, BOD_5 was determined by electronic pressure sensor in an OxiTop-C system of WTW.

3. Results and discussion

3.1. Comparison between TANFLOC and alum effectiveness

As a first approach to the importance of this new flocculant agent, a general test comparing alum effectiveness has been carried out. Raw water has been treated with 100 ppm of each product in a standard Jar-test procedure, which consisted of 100 rpm for 2 min and 30 rpm for 20 min, 1-hour settling and samples collected from the supernatant clear surface. Both products have demonstrated a high level in clarifying, almost the same in turbidity removal, COD and BOD_5. In the case of $KMnO_4$ oxidability (another measure of organic matter) TANFLOC has revealed a very slight enhancement compared with alum. These results may be seen in Fig. 1.

Using this new tannin-based flocculant may encourage attending to three main aspects:

a) Natural origin of TANFLOC makes it more affordable and more available than alum, as it can be synthesized directly *in situ*.
b) pH adjustment is not needed in TANFLOC water treatment, so reagent saving is guaranteed.
c) Several health considerations may be done referring to Alzheimer's disease and alum [32]

3.2. Jar-test parameters

Two main parameters have been varied in order to optimize Jar-test procedure. This assay consists of two stages: a rapid mixing stage (100 rpm, 2 min) which was kept as there are evidence that it would enhance the final result [28] and then another slow mixing stage, which was the target of the study. Exhausting TANFLOC effectiveness is not favorable in order to evaluate the importance of these parameters, so an intermediate dosage (20 ppm) was selected to work on.

3.2.1. Agitation speed

Agitation speed was varied between 10 and 50 rpm for a fixed period of 10 min. Turbidity removal results are shown in Fig. 2. As it

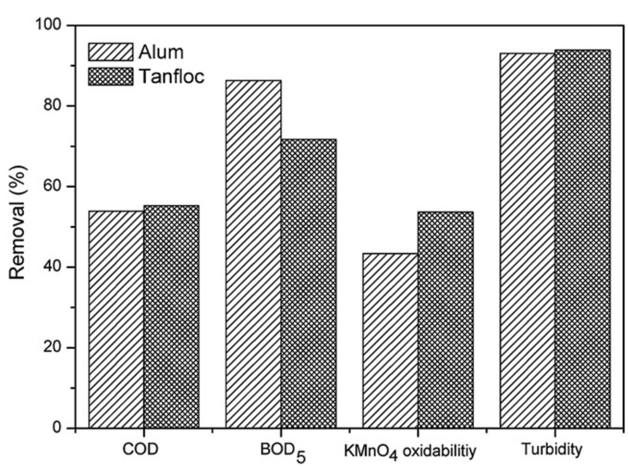

Fig. 1. Effectiveness comparison between TANFLOC and alum.

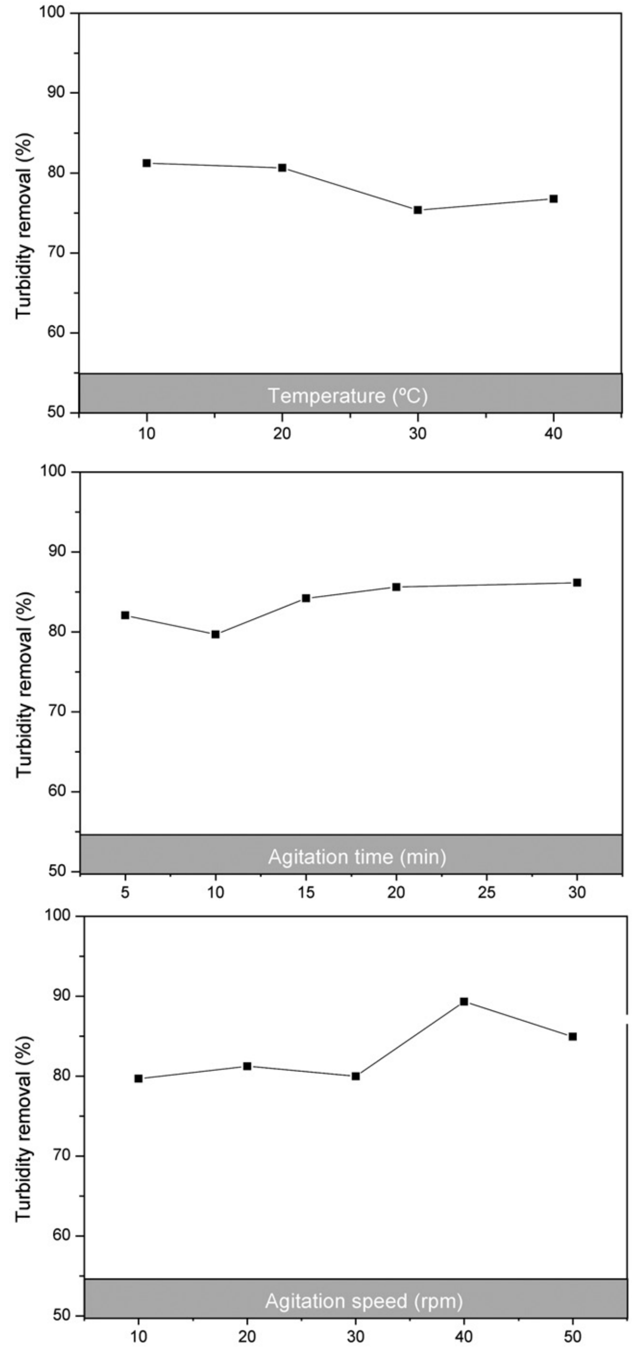

Fig. 2. Influence of temperature, agitation time and agitation speed.

can be appreciated, no high differences are found affecting this parameter. Turbidity removal varies between 80 and 90%. It keeps rather constant, but a slight improvement is observed when increasing agitation speed from 30 to 40 rpm. Stirring speed is important from the point of view of helping flocs to be formed [33] so 40 rpm value was selected as optimum one.

3.2.2. Agitation time

A 40 rpm speed was applied then in a time-variable assay. Agitation time was varied from 5 to 30 min. Fig. 2 shows an almost linear variation of effectiveness in turbidity removal. This parameter seems to be not so important, as turbidity removal varies between 80 and 90% too. 30 min was selected as an average value in order to complete the Jar-test procedure that would be used in the whole investigation: 100 rpm, 2 min plus 40 rpm, 30 min.

3.3. Temperature influence

Temperature has been evaluated as a factor in the coagulation/flocculation process. The reason why it has been introduced in this study has to do with seasonal variation; temperature of effluents may be rather different in summer than in winter; or affecting to lakes or ponds where temperature may be raised up. But temperature is also important in order to extrapolate the present results to other similar effluents, such as industrial ones, which may come into the treatment plant with very different conditions.

As it can be seen in Fig. 2 as well, temperature does not affect the effectiveness of the process. By varying temperature from 10 to 40 ºC no enhancement or worsening in turbidity removal is observed. Hence, TANFLOC may be an effective coagulant/flocculant agent even in the case of thermal-contaminated waters.

3.4. Operating parameters and treated water quality

In order to characterize treated water, several parameters that have to do with flocculation and sedimentation process have been evaluated.

3.4.1. Dosage influence

Assays with dosage variation have been carried out. Flocculant dosage has been varied between 0 and 150 ppm. As it can be appreciated in Fig. 3, turbidity removal increases quite quickly with flocculant dosage. 80%-effectiveness is achieved rather fast, with no more than 40 ppm of TANFLOC. Almost a total turbidity removal appears with dosages around 100 ppm. These results are quite competitive to those reported by other researchers. For example, Sansalone and Kim [34] has recently used up to 150 ppm of alum ($Al_2(SO_4)_3.18H_2O$) and up to 100 ppm of iron chloride ($FeCl_3$) to achieve a turbidity reduction of 75% in a similar municipal wastewater.

3.4.2. Sludge production and suspended solids removal

Sludge production is an important task in order to evaluate efficiency in coagulation/flocculation process [35]. It may be as low as possible, and sludge volume may be reduced as well. Due to this fact, aluminium-induced coagulation usually is said to be a high-sludge production process, so it became a disadvantage [36]. In the case of TANFLOC, sludge production, sludge volume and the relationship between these two parameters, which is called Sludge Volumetric Index (SVI), have been determined. SVI is defined by Eq. (1):

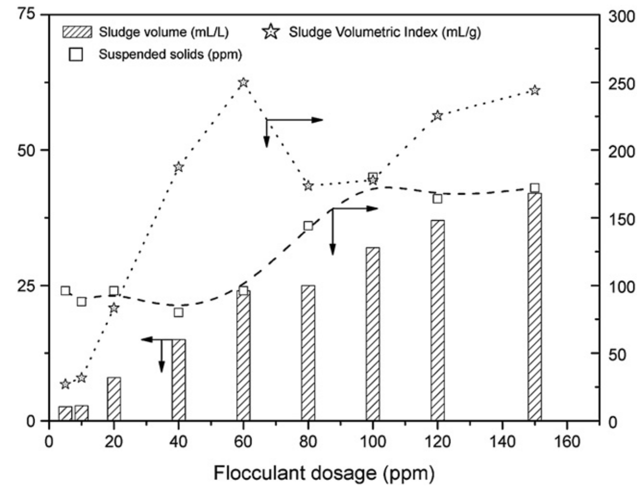

Fig. 4. Solid and sludge production variables.

$$SVI = \frac{V_s}{W_s} \quad (1)$$

where

V_s is the volume that is occupied by the sludge (mL) and;

W_s is the sludge mass (g).

As it can be appreciated in Fig. 4, the three magnitudes increase as flocculant dosage becomes higher. Suspended solids and sludge volume increasing have a less steep slope than SVI. From 80 ppm and ahead, flocculation capacity of TANFLOC seems to be less efficient, and a sludge compression seems to appear as SVI decreases. This fact is rather normal in sedimentation process [37]. These SVI values are quite interesting because they are rather lower than others, as reported by Fernández-Leborans and Moro [38] where almost every SVI value was over 150 mL/g. In addition, several disadvantages linked to alum or iron salts are avoided [39].

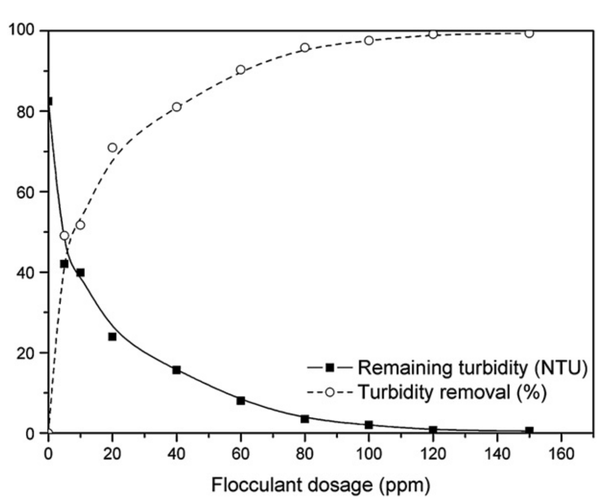

Fig. 3. General turbidity removal evolution with flocculant dosage.

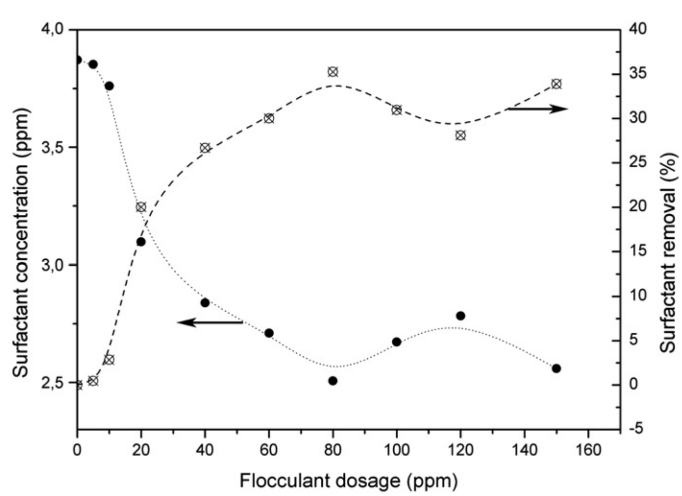

Fig. 5. Surfactant removal assay.

3.4.3. Anionic surfactant and polyphenol removal

3.4.3.1. Anionic surfactants. Surfactant dumping into environment represents a harmful and noxious practice. They may be useful and needed compounds, but they are also considered dangerous and non-desirable substances because of their impact on water animal and vegetal life. The main aspects in which surfactants modify on environmental equilibrium involve groundwater and lakes pollution, pharmaceutical product binding (so pollution activity of these kinds of chemical compounds is considerably increased), animal and human toxicity and biopersistance [40]. These are the main reasons it has been evaluated anionic surfactant removal by this tannin-based flocculant.

As it can be seen in Fig. 5, TANFLOC reaches to remove almost 30% of anionic surfactants, surely due to surfactant-turbidity adsorption and further turbidity removal. This removal tends to be constant since 60–80 TANFLOC ppm dosage and ahead, as no improvement is observed with the highest dosages.

3.4.3.2. Polyphenols. As TANFLOC is a modified tannin extract, it has been determined as tannin content which remains in equilibrium after assay. Fig. 6 shows residual polyphenol level in water. As it can be seen, it keeps reasonably constant, or with a very slight decreasing, until 60 ppm TANFLOC dosage is reached. From then and above, tannin content begins to increase. This is surely due to the fact that efficiency of TANFLOC becomes lower since this point, so a fraction of flocculant remains in water without being removed by flocculation. Anyway, tannin content in water is much lower than other values found, e.g. in tea beverage [41].

3.4.4. Organic matter removal

Finally, COD and BOD_5 removal has been analyzed with this treatment. As it can be seen in Table 1, not so high levels of organic matter are found in raw water (210 and 130 O_2 ppm for COD and BOD_5 respectively). However, a quite decreasing in both parameters is achieved with a reasonably low flocculant dosage. Fig. 7 shows a maximum COD removal around 60 TANFLOC ppm; and a maximum BOD_5 removal around 20 TANFLOC ppm. Similar ranges of COD removal have been reported by Guida et al. [42]. Biodegradability (understood as the relationship between COD and BOD_5) is rather constant and comprised inside the range of 0.5–0.7, which represents a quite high value if compared with other types of wastewater [43].

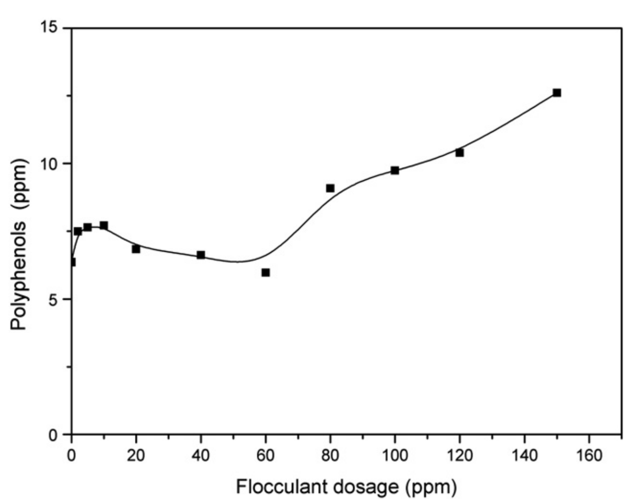

Fig. 6. Residual polyphenol assay.

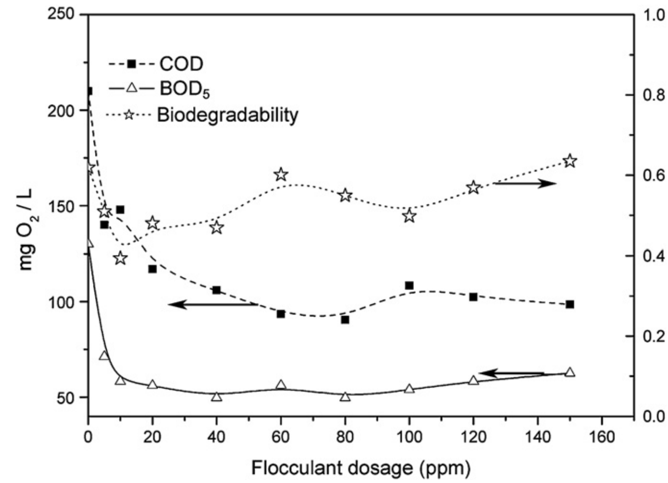

Fig. 7. COD, BOD_5 and biodegradability evolution with flocculant treatment.

4. Conclusions

Several conclusions may arise out from the present investigation:

- New tannin-based flocculant, TANFLOC, can be used as a coagulant/flocculant agent in urban wastewater treatment. Jar-test procedure may be adjusted to two agitation stages: one with rapid stirring (100 rpm) for 2 min followed by another one with slow stirring (40 rpm) for 30 min. No temperature influence has been reported.
- Effectiveness of TANFLOC is comparable in all senses with alum ability for removing BOD_5, COD and turbidity.
- Up to 80% of turbidity removal is achieved with around 40 ppm of TANFLOC, so low dosages of flocculant are quite effective in water treatment.
- Sludge production is reasonably within normal ranges, and presents no aluminium or iron salts disadvantages.
- Up to 30% of anionic surfactant is removed with TANFLOC treatment, and no excessive polyphenol content is observed in treated water.
- A reasonably COD and BOD_5 reduction is obtained by TANFLOC treatment. Water biodegradability may be found to be in the range of 0.5–0.7.

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